1-20161/A/CONT 4
July 1999

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of	:	
Bernhard Müller	:	
Serial No. 08/801,327	:	Group Art Unit 1751
Filed February 18, 1997	:	
For FIBRE REACTIVE ANTHRAQUINONE DYES, PROCESS FOR THEIR PREPARATION AND THE USE THEREOF	:	Examiner M. Einsmann

DECLARATION

I, Bernhard MÜLLER, a citizen of Germany, residing at 79588 Efringen-Kirchen, Im Mühlegrund 20, hereby declare:

That I was awarded the degree of a Doctor of Natural Science (Chemistry) of the University of Heidelberg (Germany) in 1990;

That I have been employed by Ciba Specialty Chemicals, Basel, as a chemist since 1990 and presently hold the position of a Research Chemist in the Colors Department;

That I have been engaged in the field of dyestuffs for Ciba Specialty Chemicals since 1990;

That based on the above education and experience, I consider myself an expert in the field of dyestuffs.

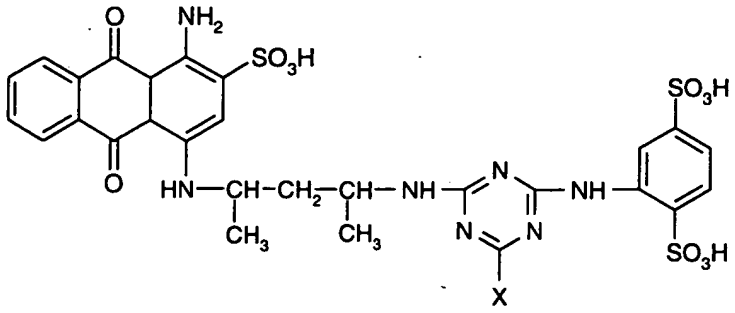
I, Bernhard MÜLLER, declare that the procedures described below were carried out under my direction and supervision;

That I am submitting herewith the following exact report of the procedures mentioned below.

---

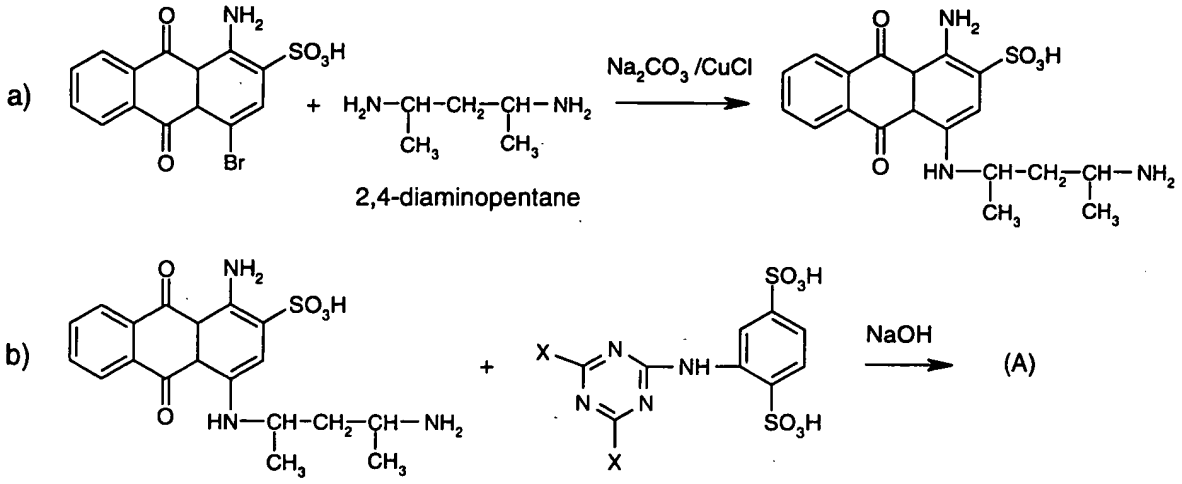
1. Introduction

Following the instructions disclosed in Examples 36 and 73 of GB-A-2 034 731, the prior art dyes of formula (A)



(A),

wherein X is F (Example 48 of GB-A-2 034 731) or Cl (Example 74 of GB-A-2 034 731) can be obtained according to a) and b) of the following scheme, wherein X has the meanings indicated above:



GB-A-2 034 731 gives **no information** on the preparation of 2,4-diaminopentane used in the Ullmann-condensation outlined in a) of the scheme, supra.

2. Preparation of 2,4-diaminopentane according to procedures known in the art

2.1 Catalytic hydrogenation of pentanone-2,4-dioxime

Pentanone-2,4-dioxime was subjected to catalytic hydrogenation according to procedures A, B and C, the particulars (catalyst, pressure, work-up) and the results of which are given below:

2.1.1 Procedure A:

catalyst: methanol / ammonia / Raney-Nickel

H_2-pressure: 150 bar

work-up: the catalyst was removed from the crude reaction mixture by fitration; volatile components were removed by distillation; the oily residue was fractionated into two fractions by distillation each of which was analysed by gas-chromatography

result: numerous products were detected in each fraction

2.1.2 Procedure B:

catalyst: ethanol / HCl / platinium oxide

H_2-pressure: 150 bar

work-up: the catalyst was removed from the crude reaction mixture by fitration; volatile components were removed by distillation; the residue was made alkaline and extracted with ethyl acetate; the isolated organic phase was dried and evaporated; the residue was analysed by gas-chromatography

result: numerous products were detected

2.1.3 Procedure C:

catalyst: acetic acid / platinium oxide

H_2-pressure: 150 bar

work-up: the catalyst was removed from the crude reaction mixture by fitration; volatile components were removed by distillation; the residue was made acidic by the addition of HCl and stored at 0°C (no crystallisation occured); then the acidic residue was made alkaline by the addition of sodium hydroxide and extracted with ethyl acetate; the isolated organic phase was dried and evaporated; the residue was analysed by gas-chromatography

result: numerous products were detected

2.2 Reductive amination of 2,4-dioxopentane (acetylacetone)

Acetylacetone was added to a saturated solution of ammonia in methanol and the mixture obtained was subjected to catalytic hydrogenation under the following conditions:

catalyst: Raney-Nickel

H_2-pressure: 150 bar

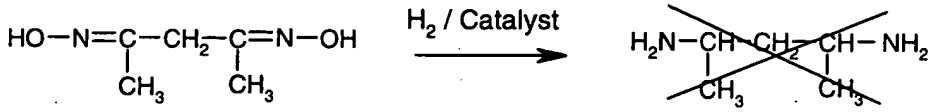
temperature: 50°C

work-up: the catalyst was removed from the crude reaction mixture by fitration; volatile components were removed by distillation; the oily residue was fractionated into two fractions by distillation each of which was analysed by gas-chromatography

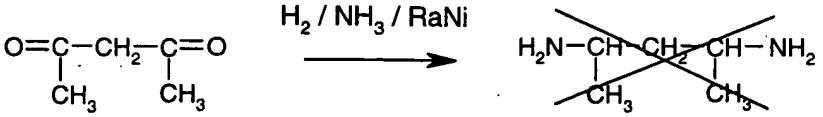
result: no product was detected (2,2-dimethylpropane-1,3-diamine was used as an internal reference);
furthermore no blue colouring was observed once the isolated fractions were subjected to an Ullmann-condensation with 1-amino-4-bromoanthraquinone-2-sulfonic acid according to a) given in the scheme of paragraph 1. (however, the reaction mixture turned blue spontaneously when 2,2-dimethylpropane-1,3-diamine was used as a reference in an Ullmann-condensation)

3. Conclusion:

a) A uniform reaction product was not obtained by catalytic hydrogenation of pentanone-2,4-dioxime under the conditions outlined in paragraph 2.1 as revealed by gas-chromatography. Schematically this is summarised below:



b) Reductive amination of 2,4-dioxopentane did not yield 2,4-diaminopentane as revealed by gas-chromatography using 2,2-dimethylpropane-1,3-diamine as an internal reference. This conclusion is further supported by the fact that no blue reaction products were obtained by Ullmann-condensation. Schematically this is summarised below:



I, Bernhard MÜLLER, hereby declare:

1. That based on my education and experience, I consider myself an expert in the field of dyestuff preparation;
2. That the procedures described, supra, and the conditions thereof are known in the art of amine preparation and are typically applied for that purpose;
3. That the results show that 2,4-diaminopentane was not obtained by the procedures described, supra.

I, Bernhard MÜLLER, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 Title 18 of the United States Code and that such wilful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 7th day of July 1999

Bernhard Müller

Bernhard MÜLLER